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SULFUR-ISOTOPE SEPARATION BY DISTILLATION*

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ABSTRACT

Sulfur-isotope separation by low-temperature distillation of hydrogen sulfide was studied in an 8-m, 25-mm diameter distillation column. Column temperature was controlled by a propane-propylene heat pipe. Column packing HETP was measured using nitric oxide in the column. The column was operated at pressures from 45 to 125 kPa. The relative volatility of S-32 vs. S-34 varied from 1.0008 to 1.0014.

INTRODUCTION

Since cryogenic distillation is an efficient means of separating isotopes of carbon (ref.1), nitrogen, and oxygen (ref.2), it was considered for sulfur isotope separation. Hydrogen sulfide was chosen for initial work because it is the lowest boiling sulfur compound, and because hydrogen is essentially monoisotopic. The overall isotopic separation in a distillation column can be accurately measured and converted to the basic vapor-liquid effect, relative volatility.

EXPERIMENTAL RESULTS

The normal boiling point of H_2S is 212.5 K. A convenient way to control a distillation column in this temperature range is to use a heat pipe condenser, a secondary refrigerant bath under a helium atmosphere cooled in turn by liquid nitrogen (ref.3). It is necessary that the secondary refrigerant remain liquid at the LN_2 temperature of 75.4 K in Los Alamos (elev. 2200 m). The eutectic mixtures of both C_3H_8-C_3H_6 and C_3H_8-C_2H_6 (ref.4) satisfy this condition. A 1:1 propane-propylene mixture was used in the present work.

The distillation column was built of stainless steel tubing and filled with stainless steel packing (4 mm dia. Propak). The column is 25 mm diameter with an active packed length of 8.1 m. Additional sampling points are located at 2.9 m and 5.2 m from the top of the packing. The entire column is enclosed in a vacuum jacket and is surrounded by multiple layers of aluminized mylar for thermal insulation. Gaseous boilup was provided by electric heaters at the column bottom; liquid reflux was maintained using the heat pipe.

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Since a distillation column multiplies a basic vapor-liquid separation, it is necessary to know how many stages or "plates" are in the column. Nitric oxide, which has large and well-known isotopic separation factors, was used to measure the number of plates in the column. Results varied somewhat, but an average number of plates was 115, resulting in a height equivalent to a theoretical plate (HETP) of 7.0 cm.

The column was operated at total reflux until steady-state isotopic separations were obtained (typically several days). Isotopic analyses were done by directly analyzing the H_2S in a CEC 21-621 cycloid mass spectrometer. Although the fragmentation ions of HS^+ and S^+ were present in the mass spectrum, their ratios to the parent peaks remained quite constant, and results were repeatable. Isotopic separations are given in Table 1.

TABLE 1
Sulfur isotope separation of ^{32}S vs. ^{34}S

H_2S pressure	Overall separation, A	Relative volatility, α
45 kPa	1.161	1.0013
46	1.167	1.0013
49	1.163	1.0013
55	1.168	1.0013
57	1.174	1.0014
120	1.109	1.0008
121	1.123	1.0010
125	1.121	1.0010

CONCLUSIONS

Sulfur isotopes may be separated by H_2S distillation; however, the effect is sufficiently small to make the method unpromising for practical use. Previous work on this system (ref.5) indicated a higher α, but that work involved single stage vapor-liquid equilibrium measurements with the radioactive isotope ^{35}S. Accurate measurements are much more difficult by that method.

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